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TRISTAN I - Techniques, Capabilities and Accomplishments

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ABSTRACT

Following a brief description of the TRISTAN facility, the techniques developed for on-line nuclear spectroscopy of short-lived fission products, the studies possible, and the activities studied are presented. All journal publications relating to the development of the facility and the studies carried out using it are referenced, and co-workers identified.

I. INTRODUCTION

In 1965, the Ames Laboratory research reactor became available for experiments in nuclear, materials, and chemical sciences. In 1966, the TRISTAN on-line isotope separator facility became operational, with the observation of mass-separated gaseous fission products in October. For nearly ten years, the TRISTAN facility (to be referred to below as TRISTAN I) set the stage for a series of successful spectroscopic studies and developments of techniques. On July 1, 1976, a new in-beam ion source was successfully operated which gave rise to non-gaseous fission products, thus extending significantly the coverage of short-lived neutron-rich nuclei. With this new approach, the TRISTAN facility became known as TRISTAN II.

Due to funding priorities in the Division of Physical Research of ERDA (now DoE), operation of the Ames Laboratory research reactor will cease at the end of calendar year 1977. Thus, the question of continuance of the existing research facilities at the reactor, such as TRISTAN II, arises and constitutes the underlying theme of this workshop. We are thus participating in discussions which are critical to the consideration of the future of studies possible with TRISTAN II, relocated to the High Flux Beam Reactor at Brookhaven.

There will be three talks on TRISTAN; this one, on the past achievements, both technical and scientific, of TRISTAN I; Fred Wohn will discuss the future promises of a continuance of TRISTAN II; and John Hill will speak to the studies which have been made during the short span of TRISTAN II.

Rather than repeat much of the information which has appeared in print, I would like to present a complete listing of pertinent references and discuss some of the philosophy behind the development of the capabilities and studies with TRISTAN I. The references are divided into system description and instrumentation development,[1-11] and the studies carried out,[12-48] essentially in chronological order except for the unpublished work (which, hopefully, will make its way into print). The definitive description of the system is Ref. 10.

I should like to emphasize at the outset that the development of the TRISTAN facility has been the product of the efforts of many people, and I will acknowledge their contributions now rather than at the end of my talk, to underscore my dependence upon them for the activities which resulted in the highly productive program at TRISTAN I. Table I illustrates what has not been properly appreciated up to now, in my opinion: that the number of students who received their training and did their research, compared to the number of dedicated staff (which at any one time numbered about 4 FTE's), is very large indeed.

II. TECHNIQUES ASSOCIATED WITH TRISTAN I

The initial concepts envisioned with TRISTAN were derived from two facts of the ^{235}U thermal fission process, both observable from the fission yields shown in Fig. 1. First of all, when viewing the mass yield profile, there are essentially two well-defined mass regions for study, and these regions are separated by roughly a chemical period. Secondly, when viewing the chemical yield profile, the gaseous products, Kr and Xe, both have yields close to the peak chemical yields. The accented individual chemical yields for Kr and Xe illustrate, moreover, that decay products of the high-yield gaseous activities would be produced in higher abundance through decay than by direct fission yield.

Given these facts in combination with the unique physical properties of the noble gases (high volatility and low chemical activity), the development of a system which would make available the gaseous fission products would not only be highly tractable, but also result in a scientifically significant coverage of neutron-rich activities. Coupled with these considerations is the need to concentrate on the techniques which are to be employed in looking at short-lived activities at an on-line facility. The latter effort was needed to be able to anticipate and address the procedures necessary for on-line studies of short-lived

TABLE I. List of co-workers

STAFF: 2 years or more

J. R. McConnell
F. K. Wohn
W. C. Schick, Jr.
J. C. Hill
K. L. Malaby
A. R. Landin
M. A. Cullison
J. J. Eitter
G. H. Carlson
J. C. Pacer

Less than 2 years

B. Anderberg
K. B. Nielsen
G. M. Day
A. B. Tucker
B. R. Erdal

STUDENTS:

C. L. Duke
D. Thomas
J. T. Larsen
G. H. Carlson
D. I. Haddad
J. E. Solecki
J. R. Clifford
R. J. Olson
J. W. Cook
J. K. Halbig
J. H. Norman
E. A. Henry
J. J. Eitter
J. P. Adams
M. A. Lee
J. A. Morman
R. S. Weinbeck
G. J. Basinger
L. J. Alquist
R. L. Bunting
M. D. Glascock
W. R. Western
R. L. Gill
C. J. Bischof
G. A. Sheppard
K. A. Burke
J. F. Wright

COLLABORATORS:

R. J. Hanson
H. H. Hsu
S. T. Hsue
C. L. Duke
D. M. Roberts
J. W. Layman
A. F. Voigt

VISITORS:

P. Paris
H. K. Carter
J. P. Zirnheld
J. Lin
T. A. Khan
S. Amiel
J. C. Wells, Jr.

ENCOURAGEMENT AND ADVICE:

I. Bergström
A. Kjellberg
S. Borg
G. Rudstam
R. D. Macfarlane
R. A. Naumann
S. Raman
B. J. Dropesky
G. Herrmann
M. E. Bunker
A. A. Bartlett
P. G Hansen
S. Sundell
R. A. Meyer
G. B. Holm

SPONSORSHIP:

G. L. Rogosa (AEC and ERDA)

F. H. Spedding
D. J. Zaffarano
R. S. Hansen
} Ames Laboratory and Iowa State University

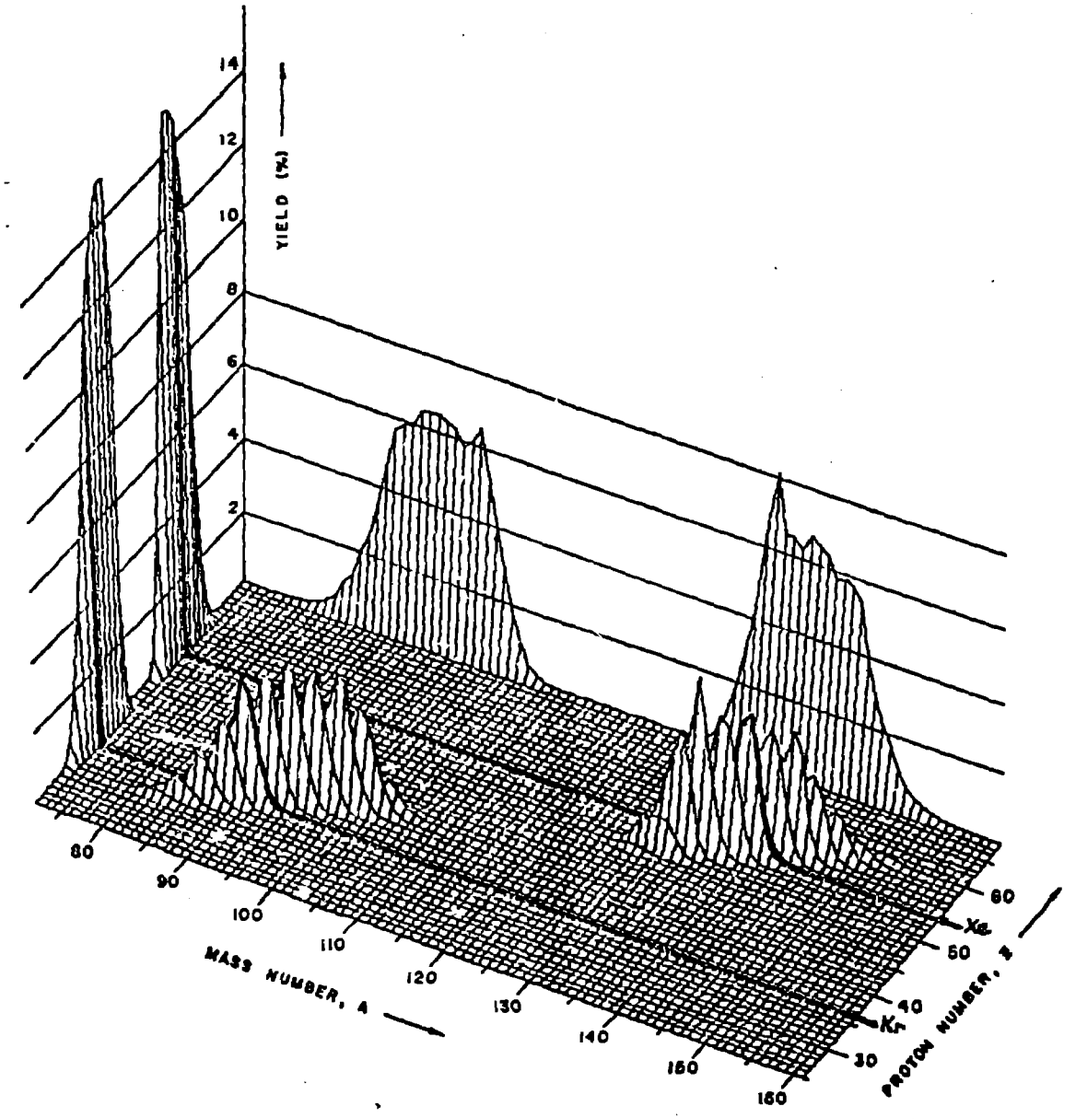

Fig. 1. Isometric view of the yields for thermal fission of ^{235}U.

nuclei where the daughter activities are also short-lived.

The resulting system configuration is shown in Fig. 2, which shows the layout of TRISTAN I as it was used from 1969 until 1976. A detailed description of the system is given in Ref. 10; for the purpose of this talk, a short description will suffice. The sample of ^{235}U (in the form of the stearate or tetra-fluoride) was placed in a neutron beam from the reactor, of nominal diameter 5 cm and flux 3×10^9 n/cm^2/sec. The sample size was nominally 10 g of ^{235}U. Operation of the sample chamber and transport line at ambient room temperature resulted in the availability of predominantly the Kr and Xe fission products, with small amounts of Br and I. The two-stage separation provided by the separator followed by the switch magnet resulted in mass separation factors which were not measurable; that is, despite an estimated detection sensitivity of 10^7 to 1, no (A+1) activity could be detected at the (A) deposit after the switch magnet. The same statement did not hold true for (A-1) contaminants; the presence of KrH^+ and XeH^+ ions represented a real contribution to the deposit under study, and resulted in a (A-1) contamination level which was significant in the higher-mass studies. The combination of lower fission yield and shorter half-life (resulting in larger loss in transport) for the high-mass products compared to the (A-1) activities, resulted in an enhancement of the KrH^+ and XeH^+ deposition rates for these studies, even though the hydride activities were typically 10^{-4} the ionic activities for the same isotope.

Despite the above problem of hydride contamination, the principal concern in the studies of short-lived fission products was that of the interference from short-lived daughter activities. Accordingly, considerable effort was made in the development and analysis of moving tape collector technology, which provided a mechanical means of discrimination between activities of an isobaric decay chain. The principle of the moving tape collector can be seen in Fig. 3. In the collection of the separated ion beam, the ions are actually imbedded in the collecting surface. If this surface should be a tape which is capable of motion, the motion will carry the deposit from one place

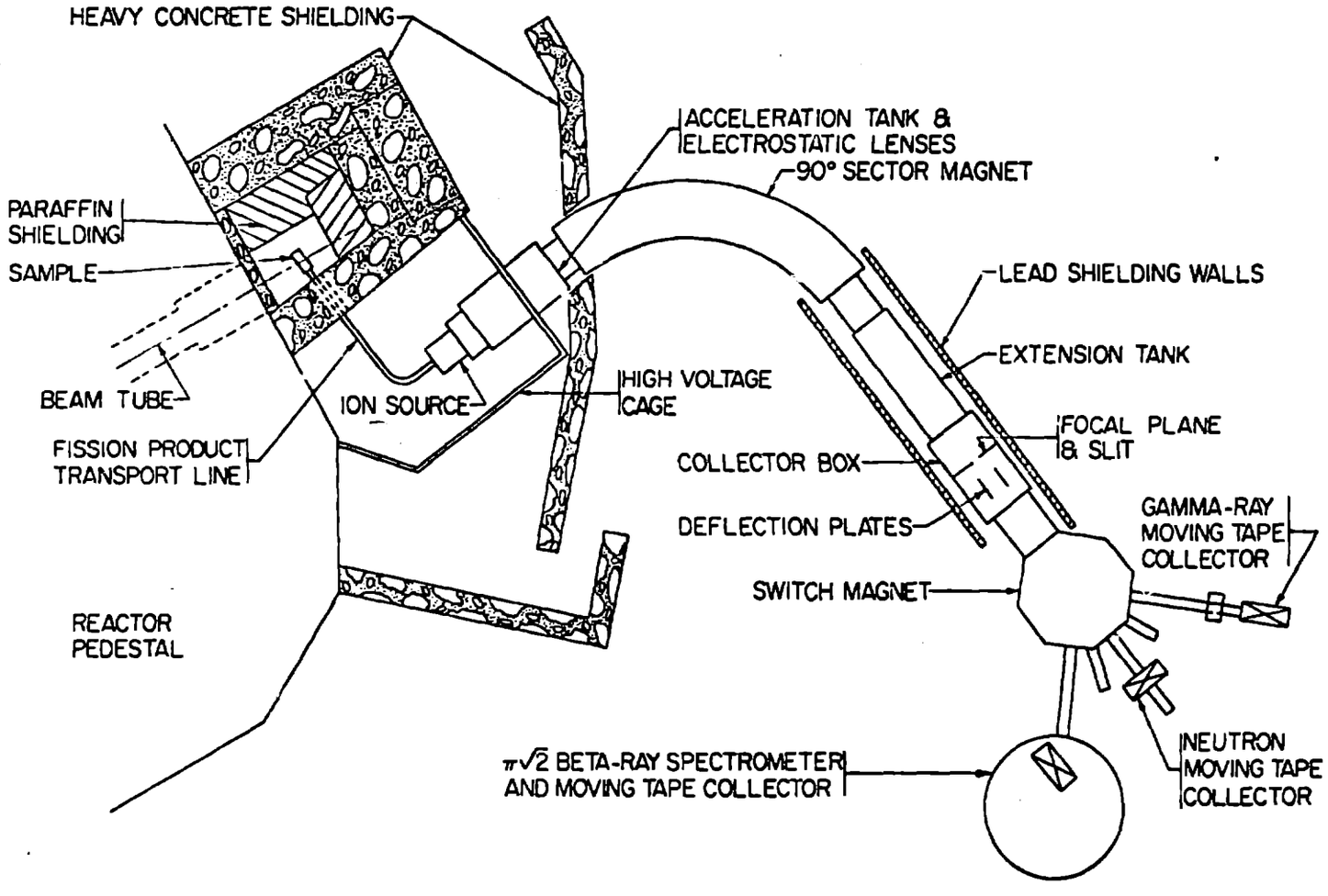

Fig. 2. Layout of the TRISTAN I facility.

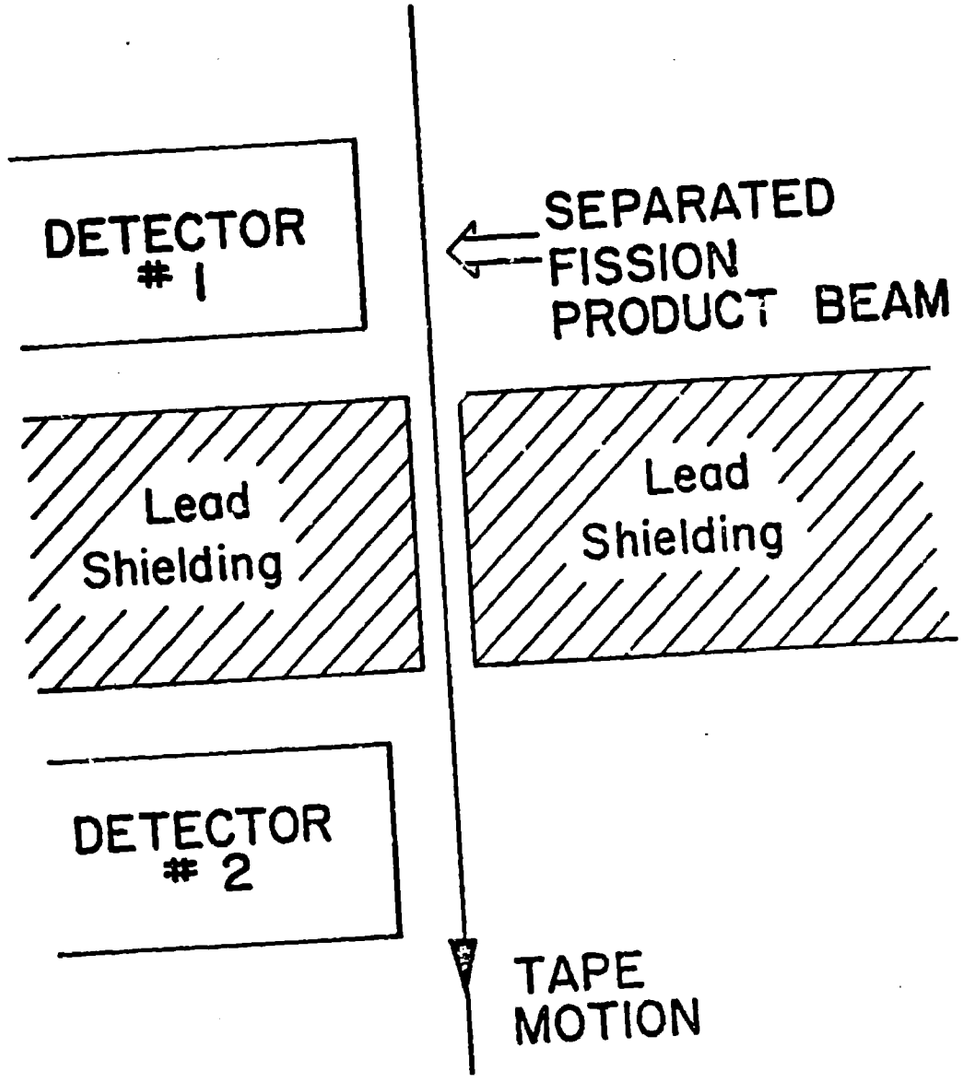

Fig. 3. Moving tape collector principle.

to another. In considering the situation pictured in Fig. 3, with the tape in continuous motion, only the shortest activities in the deposition will decay before being moved into the lead shielding and away from detector #1. Thus, detector #1 will not see the longer-lived activities in the deposit. On the other hand, by the time the deposit has moved through the lead shielding to detector #2, the shortest-lived activities have decayed away, leaving only the longer-lived activities to be viewed by detector #2. In an isobaric decay chain of a Kr or Xe separated isotope deposit, the parent decays (Kr or Xe) are generally the shortest-lived activities and the daughter activities which build in as the parent decays are longer-lived. Detector #1 can thus be made sensitive to the parent decays and detector #2 to the daughter decays by appropriate choice of tape speed. We have found that the discontinuous motion of the tape is a preferred mode of operation, with selection of deposition, delay, and data acquisition times to optimize the temporal conditions for a selected activity within the isobaric decay chain.[4]

The moving tape collector principle has been applied to all the studies carried out. The "Gamma-Ray Moving Tape Collector" shown on the layout (Fig. 2) has been designed to accommodate electron spectrometers, as well as angular correlation detectors. Separate moving tape collectors were constructed for use with a neutron spectrometer and a $\pi\sqrt{2}$ spectrometer.

Among the other techniques developed for TRISTAN, without elaboration, are: the on-line $\pi\sqrt{2}$ beta-ray spectrometer,[8] an absolute beta counter for ground-state branching,[37] and a multiple-detector angular correlation detector array.[9] In addition, beta-ray and electron spectrometers were developed for decay energy and conversion electron studies.[6,30] Taken in total, the complement of experimental techniques available at TRISTAN represented perhaps the most complete spectroscopic capability at a single experimental facility. Still, the use of the facility was mainly limited by the available manpower.

III. CAPABILITIES

While the discussion on techniques gives an indication of the capabilities which result, a few comments can be made using the help of the stylized gamma-ray spectrum and decay scheme shown in Figs. 4 and 5, respectively. Apologizing in advance for being simplistic to those in the audience who are accomplished level scheme builders, let me explain that the analysis of the spectrum shown in Fig. 4 reveals that five gamma rays are emitted in the decay of the observed activity, and that these five gamma rays can be placed into the level scheme of Fig. 5 by using the energy-sum relationships evident from the analysis. The placement of γ_2 and γ_4 can be verified, in a general study, by showing them in coincidence with γ_1 (assuming a very short lifetime for the first excited state).

The existence of the beta branches, including that to the ground state, can be inferred from the intensity imbalances for feeding and depopulating the excited states, as observed from the gamma-ray intensities. For the ground-state branch, an additional piece of information is required, the total beta decay intensity (obtainable using a 4π beta detector). As frequently happens for short-lived fission products, the decay energy may exceed the neutron binding energy for the daughter nucleus. In this case, direct neutron emission may occur after beta decay, even in competition with gamma de-excitation. This phenomenon, referred to as delayed neutron emission, is of great interest from reactor control studies and also from the point of view that such emission constitutes observable information on the nature of highly excited states in nuclei.

Then, in view of the decay scheme shown in Fig. 5, the capabilities which should be covered are: half-life determination, delayed neutron emission, decay energy, ground-state beta branching, conversion coefficient measurements (to more fully characterize the de-excitation parameters), gamma-ray spectroscopy, coincidence studies, level lifetimes, and angular correlation studies (to measure de-excitation spin sequences). All of these capabilities were available at TRISTAN. The only nuclear

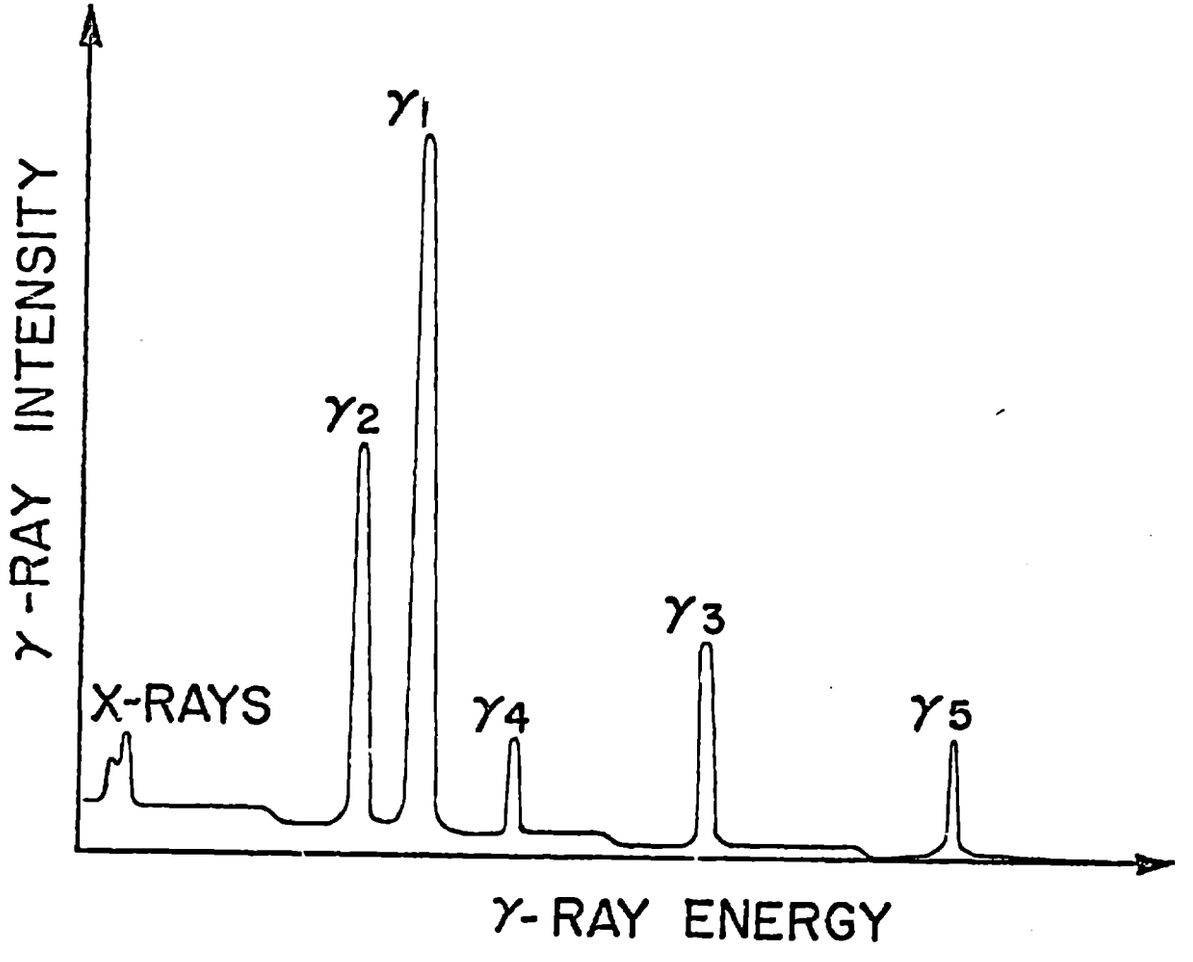

Fig. 4. A simple gamma-ray spectrum

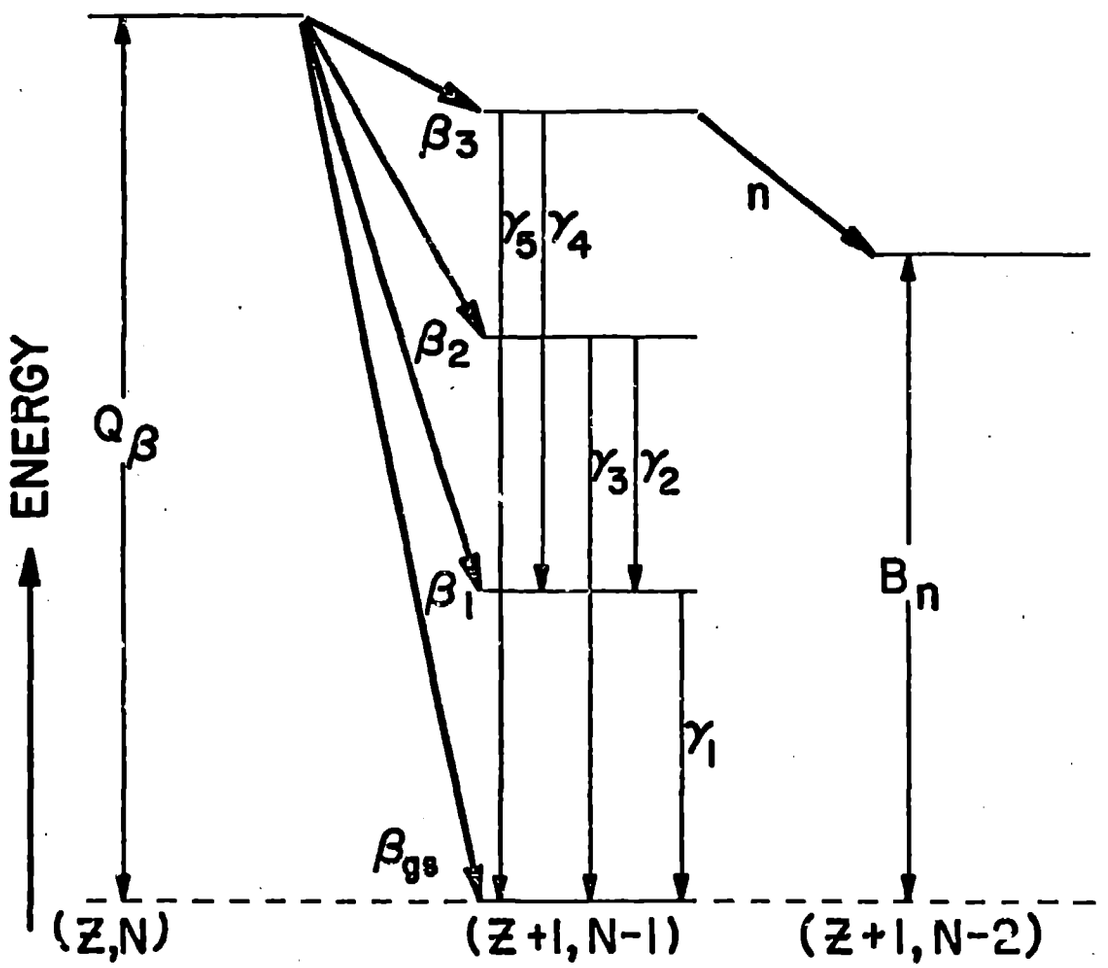

Fig. 5. Generic decay scheme of a short-lived neutron rich nuclide.

parameters not unambiguously determined with these capabilities are the spins and moments of the ground states, for which a technique employing the nuclear hyperfine interaction seems feasible and is, indeed, under development. The determination of decay energy could also be accomplished using an on-line high-resolution mass spectrometer for direct mass measurement of the ground-state. It should be noted, however, that development of the latter two techniques is considerably more involved than for those at hand, with larger manpower resources inevitably required.

A word of caution: In considering the continuance of TRISTAN II, it would be advisable to install a further capability, essentially unrelated to the techniques described thus far. This is the on-line, interactive analysis capability afforded through an expanded-memory minicomputer system. If TRISTAN I had any real shortcomings, it was in the time required to analyze the data. An interactive analysis system, on-line or off-line, would have reduced the time spent on gaseous fission products considerably.

The last capability which should be required of a general facility to study neutron-rich fission products is that of greater coverage of the available elements than just the gaseous products. Although TRISTAN II represents just that capability, I would like to unveil the fact that as far back as 1968, I had attempted to summon additional support for the facility to investigate the possibilities for increased elemental availability. Given the lack of such support, we proceeded to do the best we could with the activities available until, about 1974, it was evident that we had to invest our current effort into the developments which eventually proved successful two years later. This was at the expense of completing the possible studies with the gaseous activities, and is at least partly responsible for the unpublished status of some of the studies which were carried out.

A summary of the capabilities, as well as the studies carried out, is shown in Table II. References are provided for the activities and studies made, and the techniques employed are described in the individual cases.

TABLE II. Spectroscopic capabilities and studies at TRISTAN

Type of study	Spectroscopic technique	Activities studied	References*
$T_{1/2}$	Ge(Li) spectrum multiscaling	^{85m}Kr; ^{89}Kr,Rb; ^{90}Kr,Rb; ^{91}Kr,Rb; ^{92}Kr,Rb; ^{93}Kr,Rb,Sr; (^{94}Kr,Rb); ^{136}I; ^{137}Xe; ^{138}Xe,Cs; ^{139}Xe,Cs; ^{140}Xe,Cs; ^{141}Xe,Cs,Ba; ^{142}Xe,Cs,Ba	14, 18, 41
P_n	BF_3 long counter	^{92}Kr,Rb; ^{93}Kr,Rb; ^{141}Xe,Cs; ^{142}Xe,Cs	12, 13
Q_β	$\pi\sqrt{2}$ mag. spect.	^{85m}Kr; ^{87}Kr; ^{88}Rb; ^{89}Sr; ^{91}Sr	17, 18, 22 24, 43
	Plastic scint.-Ge(Li)	^{88}Kr,Rb; ^{89}Kr; ^{90}Kr,Rb; ^{91}Kr,Rb; ^{92}Kr,Rb,Sr; ^{93}Kr,Rb; ^{138}Xe,Cs; ^{139}Xe,Cs; ^{140}Xe,Cs; ^{141}Xe,Cs,Ba; ^{142}Xe,Cs	25, 26
β_{gs}	$\pi\sqrt{2}$ mag. spect.	^{85m}Kr; ^{87}Kr; ^{91}Sr	18, 22, 24
	4π plastic scint.	^{88}Kr,Rb; ^{89}Kr,Rb; ^{90}Kr,Rb; ^{91}Kr,Rb	37
	Ge(Li) relative activity	^{89}Kr; ^{91}Kr,Rb; ^{92}Kr,Rb; ^{138}Xe,Cs; ^{139}Xe,Cs	21, 23, 28 36, 44
ICC	$\pi\sqrt{2}$ mag. spect.	^{85m}Kr; ^{88}Kr; ^{90}Kr,Rb; ^{91}Kr,Rb,Sr; ^{92}Kr	18, 24, 32 43
	Si(Li)	^{140}Xe	30
	LEPS	^{91}Kr,Rb	32
γ	Ge(Li) and LEPS	^{85m}Kr; ^{88}Kr,Rb; ^{89}Kr,Rb; ^{90}Kr,Rb; ^{91}Kr,Rb,Sr; ^{92}Kr,Rb,Sr,Y; ^{93}Kr,Rb,Sr,Y; (^{94}Kr,Rb); ^{136}I; ^{137}I,Xe; ^{138}I,Xe,Cs; ^{139}Xe,Cs; ^{140}Xe,Cs; ^{141}Xe,Cs,Ba,La; ^{142}Xe,Cs,Ba,La; ^{143}Ba; (^{143}Xe,Cs,La); (^{144}Xe,Cs)	15, 16, 18 19, 20, 21 23, 27, 28 29, 35, 36 38, 39, 40 42, 44, 45 46, 48

TABLE II. (Continued)

Type of study	Spectroscopic technique	Activities studied	References*
$\gamma\gamma$	Ge(Li)-Ge(Li)	^{88}Kr,Rb; ^{89}Kr,Rb; ^{90}Kr,Rb; ^{91}Kr,Rb,Sr; ^{92}Kr, Rb,Sr; ^{93}Kr,Rb,Sr,Y; ^{136}I; ^{137}I,Xe; ^{138}I,Xe,Cs; ^{139}Xe,Cs; ^{140}Xe,Cs; ^{141}Xe,Cs,Ba,La; ^{142}Xe,Cs,Ba,La; ^{143}Ba	15, 16, 19 20, 21, 23 27, 28, 29 35, 36, 38 39, 40, 41 42, 44, 45 46, 48
$\gamma(t)$	LEPS-plastic scint.	^{91}Kr,Rb; ^{136}I; ^{140}Xe; ^{141}Xe	26, 31
$\gamma\gamma(\theta)$	Ge(Li)-NaI(Tl)	^{90}Rb; ^{138}Cs; ^{140}Xe,Cs; ^{142}La	9, 33, 34 47

*Activities in parentheses have been studied, but preliminary data have not been analyzed.

IV. ACCOMPLISHMENTS

An idea of the scope of the accomplishments of TRISTAN I, as well as the capabilities, may be obtained from studying Table II. There yet remain some areas of study which render the picture incomplete; in particular, the ground-state beta branching intensities are unfortunately not available for mass numbers higher than 91, from direct measurement. What studies are presented, however, represent detail which is unlikely to be improved upon for the foreseeable future. The results contained in the references are, in many cases, significant improvements over prior studies, and for other activities, are the only definitive results available.

Another way of summarizing the accomplishments from the use of TRISTAN I is to present the nuclei studied on a portion of the chart of nuclides. This is done in Figs. 6 and 7, in combination with the fission chain yield and an indication of the approximate half-life regions. The shaded boxes represent the activities studied at TRISTAN I. It is obvious that there are many high-yield fission products not studied at TRISTAN I, and that the as yet unstudied regions represent a challenge for years to come. It is heartening that an approach such as TRISTAN II promises to fill in the gaps of our knowledge of short-lived nuclei produced in fission, given the resources and manpower for the near-term future.

To conclude this brief, yet comprehensive, review of TRISTAN I, I would like to inject a personal note. It was my great pleasure and honor to have been a part of the scientific strides made through the development of this facility. I appreciate especially the willing and effective support I received from my co-workers during the time of development and operation of TRISTAN. It saddens me to see the work halted, however momentarily it may turn out to be, in response to fiscal pressures which are imposed just at the instant that the facility is responding fully to its potential. I can only hope that the value of the past efforts will continue to be realized by the community so that the potentials which have been built up are not forfeited.

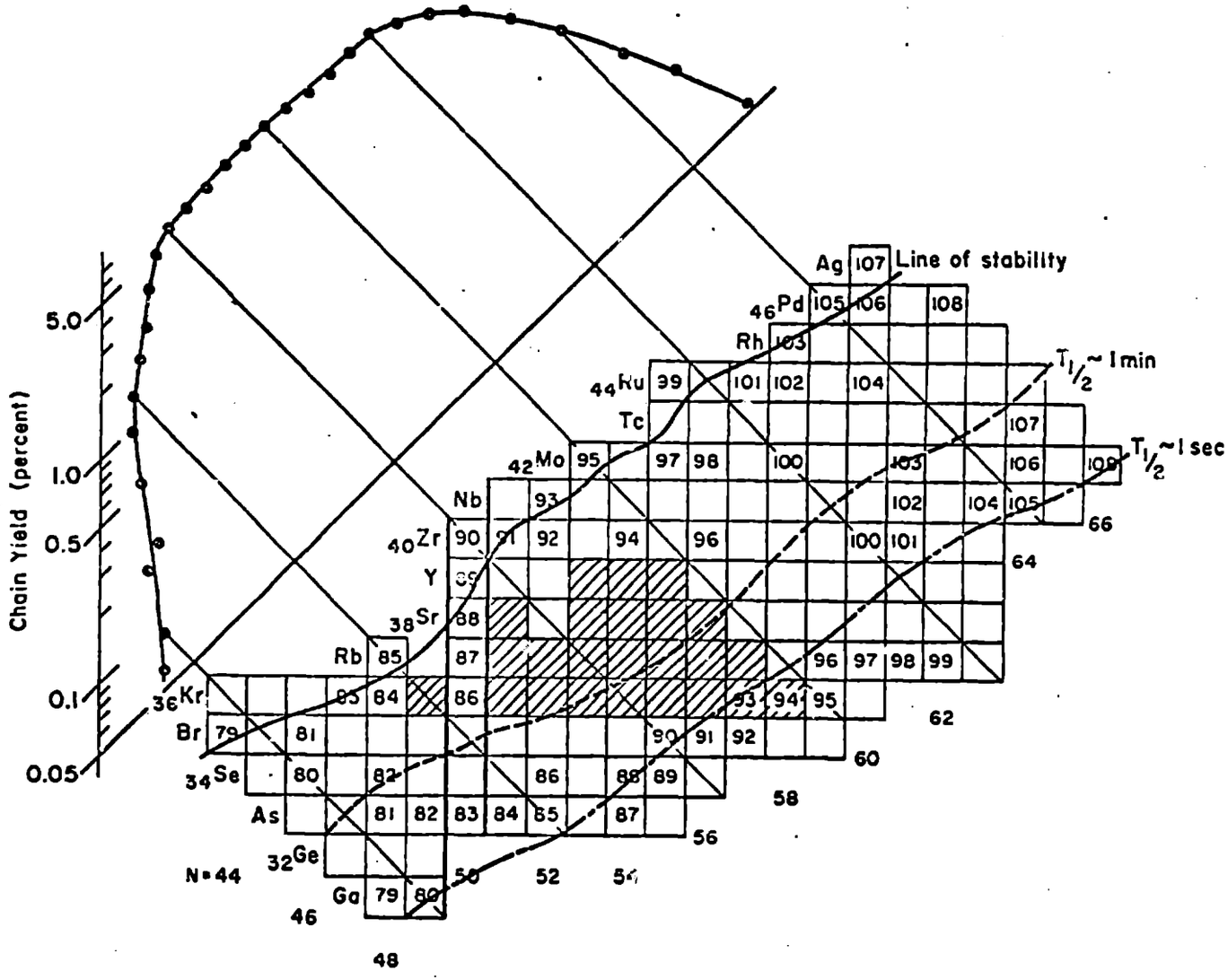

Fig. 6. Partial chart of nuclides showing low-mass fission product region. Mass chain yields are also indicated. The shaded boxes are decays observed at TRISTAN I.

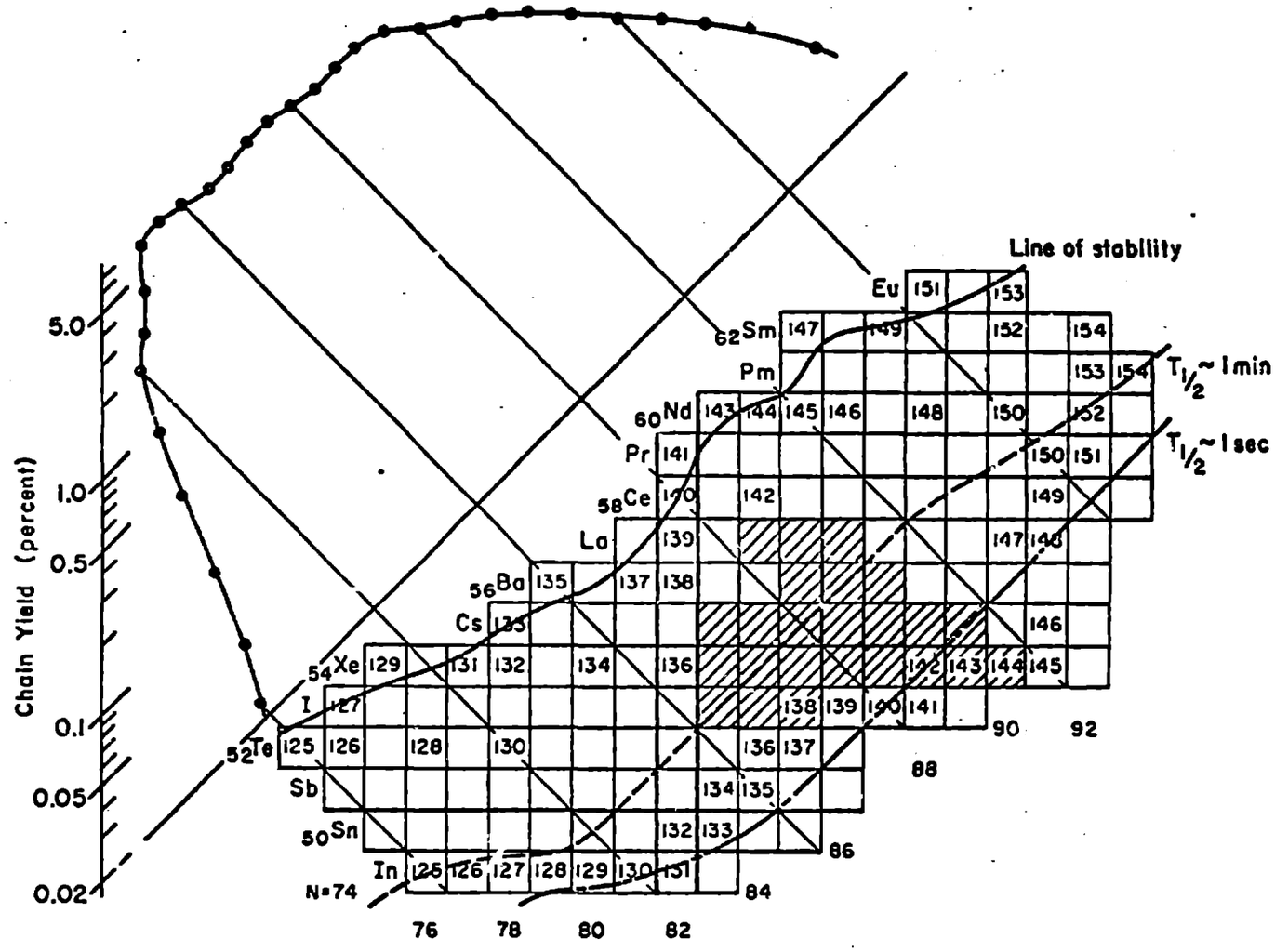

Fig. 7. Partial chart of nuclides showing high-mass fission product region. The shaded boxes are decays observed at TRISTAN I.

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IN REPLY
REFER TO: Q-14-377
MAIL STOP: 560

February 15, 1978

Dr. Robert E. Chrien
Department of Physics
Brookhaven National Laboratory
Upton, L.I., NY 11973

Dear Bob:

With apologies for tardiness, I am sending you the manuscript for my talk presented at the Ames-BNL Workshop on ISOL Systems.

I would be interested in knowing the outcome of the considerations to locate TRISTAN at the HFBR. My last talks with the Ames group indicated that BNL was willing if DoE provides adequate support. Has there been any firm commitment?

I look forward to receiving the proceedings of the workshop, and hope my contribution alone has not caused a delay in their publication.

Sincerely yours,

W. L. Talbert, Jr.

WLT:af
Enclosure: manuscript
C: [redacted]
W. L. Kirk, Q-DO/572
W. L. Talbert, Jr., Q-14/560
Q-14 file/560

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